

Tempered relaxation with clustering patterns

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Abstract

This work is motivated by the relaxation data for materials which exhibit a change of the relationship between the fractional power-law exponents when different relaxation peaks in their dielectric susceptibility are observed. Within the proposed framework we derive a frequency-domain relaxation function fitting the whole range of the two-power-law dielectric spectroscopy data with independent low- and high-frequency fractional exponents γ and $-\alpha$, respectively. We show that this effect results from a contribution of different processes. For high frequencies it is determined by random stops and movement of relaxing components, and the low-frequency slope is caused by clustering in their temporal changes.

Keywords: Tempered α -stable process, Fractional two-power relaxation, Compound subordination

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1. Introduction

The stochastic approach based on subordination of compound random processes for description of the non-exponential relaxation phenomena within the anomalous diffusion framework shows itself as one of the most useful mathematical tools [1]. Its recent success allows one to get the fractional two-power dependency commonly observed in relaxation of complex materials (glasses, liquid crystals, polymers, biopolymers and so on). According

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to this approach, the well-known Havriliak-Negami (HN) relaxation appears due to a compound subordination by two independent random processes. One of them is connected with a coupling between the very large jumps in physical and operational times, and another accounts for the amount of time when a relaxing entity does not participate in motion. In fact, both of them are described by means of a skewed α -stable process and its inverse. Although, from one side the results help one to understand better the complex nature of the physical mechanisms underlying the power-law relaxation responses, at the same time they raise a number of problems to the theory of relaxation. In particular, any α -stable process has even no the first moment. In real situations there are sufficiently many factors truncating the distribution of α -stable processes [2, 3, 4, 5, 6]. This leads to the tempered α -stable processes, and they have all the moments. As it is shown in [7], from the subordination by the inverse tempered α -stable process one can derive the tempered diffusion equation and the relaxation function describing the Debye (D), Cole-Cole (CC) and Cole-Davidson (CD) types of relaxation. The tempered diffusion has a transient character, i. e. a crossover from subdiffusion at small times to normal diffusion at long times. The transient subdiffusion has impact on kinetics of magnetic bright points on the Sun [8] and has been observed in cells and cell membranes [9, 10, 11]. Physical arguments for appearance of such effects are that subdiffusion is caused by traps, but in a finite system there is a given maximal depth of the traps (maximal waiting time) truncating their power-law waiting time distribution in such a way that beyond the maximal waiting time the diffusive behavior of the complex system tends to normal. Note also that the truncation of waiting times demonstrates features of weak ergodicity breaking in motion of lipid granules [12]. The above mentioned relaxation functions are only partial cases of the more universal HN law (see Fig. 1). Let us mention at this point that the original HN function fits [13, 14] the fractional power-law dependence observed in large part of the dielectric susceptibility $\chi(\omega)$ data:

$$\begin{aligned} \Delta\chi(\omega) = \chi(0) - \chi(\omega) &\propto (i\omega/\omega_p)^m && \text{for } \omega \ll \omega_p, \\ \chi(\omega) &\propto (i\omega/\omega_p)^{n-1} && \text{for } \omega \gg \omega_p \end{aligned} \quad (1)$$

with $0 < m < 1$ and $0 < 1 - n < 1$, the low- and high-frequency exponents, respectively. It has to be stressed the HN exponents fulfill the following relation $m > 1 - n$. Here ω_p denotes the loss peak frequency, $(i\omega)^\lambda$ means $(i\omega)^\lambda = |\omega|^\lambda \exp(i\lambda\pi \operatorname{sgn}(\omega)/2)$ and $i = \sqrt{-1}$. The data characterized by the opposite relation $m < 1 - n$ (the diagram part of Fig. 1 below

the line CC) cannot be interpreted in terms of this function, since its origins, found within the fractional Fokker-Planck equation [15] and continuous time random walk [16, 17] approaches, cannot yield this relation. The corresponding time-domain description uses the notion of a response function $f(t) = -d\phi(t)/dt$ (negative time derivative of the relaxation function $\phi(t)$), exhibiting the following power-law asymptotics

$$f(t) \propto \begin{cases} (t/\tau_p)^{-n} & \text{for } t \ll \tau_p, \\ (t/\tau_p)^{-m-1} & \text{for } t \gg \tau_p, \end{cases}$$

where $\tau_p = 1/\omega_p$ is the characteristic relaxation time, determined by the loss peak frequency. Here is useful to mention that an ideal capacitor with an impedance of the form $1/(i\omega C)$ does not exist in nature, and as a rule, dielectric materials exhibit a more realistic fractional behavior $1/(i\omega C)^\alpha$ with $\alpha < 1$ [18].

The main open question, arising here, is whether it is possible to describe such a general relaxation picture (in the sense of arbitrary exponents m and $1 - n$) by the tempered scheme. It would be interesting and important for understanding of the relaxation mechanism, especially when a change of the relationship between the power exponents from $m > 1 - n$ to $m < 1 - n$ is observed in one dielectric system for different dielectric susceptibility peaks. As we see, the same material can have both the $m < 1 - n$ and $m > 1 - n$ relaxation patterns under different temperature/pressure conditions, and a model, in which this passage via model parameters is possible, is required. It would be also of a great importance to find a single analytical form of the relaxation function fulfilling such requirements (see Fig. 1 and Table 1).

The purpose of this Letter is to shed light on the character of relaxation processes in such materials. To reach this goal we present a new scenario of subordination based on finite-moment random processes that extend the theory of tempered relaxation [7]. The basic idea presented here also starts with a compound subordination of two random processes independent of each other. One of them is the inverse tempered α -stable process but the another one is the undershooting process with index $0 < \gamma < 1$ [1, 19].

The Letter is organized as follows: at first, we briefly define the primary random processes and formulate a modification of their subordination representation. Next, within this approach we derive the frequency-domain relaxation function in an analytical form. In the last section the results of our study are discussed as applied to relaxation phenomena.

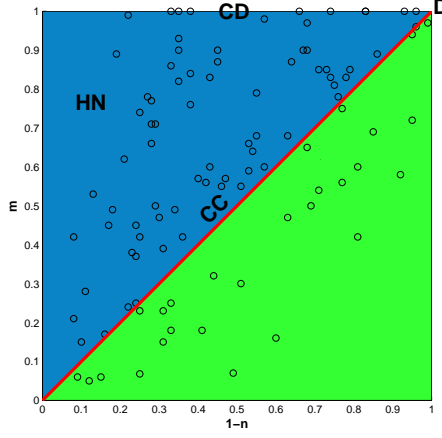


Figure 1: (Color online) Relaxation diagram positioning different laws of relaxation. The exponents m and $1 - n$ focus on declination of the imaginary susceptibility $\chi''(\omega)$ for low and high frequencies. The circles are experimental points (for various materials) taken from the book [14].

Table 1: The relation between exponents m and $1 - n$ for some experimental data (see also [14]).

Material	m	$1 - n$	Comments
Polyvinylidene fluoride	0.07	0.49	α -peak, $m < 1 - n$
Polyvinylidene fluoride	0.53	0.13	β -peak, $m > 1 - n$
Polyvinylidene fluoride	0.05	0.12	γ -peak, $m < 1 - n$
Nylon 610	0.6	0.57	$T = 483$ K, $m > 1 - n$
Nylon 610	0.3	0.51	$T = 413$ K, $m < 1 - n$
Nylon 610	0.18	0.41	$T = 373$ K, $m < 1 - n$
Glycerol	0.55	0.46	$P = 3.1$ kb, $m > 1 - n$
Glycerol	0.5	0.69	$P = 4.4$ kb, $m < 1 - n$

2. Tempering and coupling in the compound subordination

The tempered α -stable process [20, 21] is characterized by the following Laplace image of its probability density function (pdf)

$$\tilde{f}_\alpha(u) = \exp(\delta^\alpha - (u + \delta)^\alpha) , \quad (2)$$

where the stability parameter $0 < \alpha \leq 1$ and the tempering parameter $\delta \geq 0$ are constants. If δ equals to zero, the tempered α -stable process becomes simply α -stable. In other words, the parameter δ provides just a truncation of the ordinary, long-tailed totally skewed α -stable distribution. The truncation leads to the random process having all moments finite. Formula (2) describes probabilistic properties of the tempered process in terms of the internal (operational) time. Its inverse process may be used as a subordinator. The pdf $g_\alpha(\tau, t)$ of the subordinator depends on the real physical time and describes the first passage over the temporal limit t . Its Laplace transform reads

$$\begin{aligned} \tilde{g}_\alpha(\tau, u) &= -\frac{1}{u} \frac{\partial}{\partial \tau} \tilde{f}(u, \tau) = \\ &= \frac{(u + \delta)^\alpha - \delta^\alpha}{u} \exp(-\tau[(u + \delta)^\alpha - \delta^\alpha]) . \end{aligned} \quad (3)$$

The inverse tempered α -stable process accounts for motion alternating with stops so that the temporal intervals between them are random and with heavy tails in density. The main feature of the process is that it occurs only for small times [6].

If the γ -stable process is subordinated by its inverse, the compound subordinator behaves 1-similar (proportional to time t) for long times [22]. Such a subordinator does not, however, lead to any fractional two-power relaxation dependency like the HN one. Therefore, we consider a more complex subordination, where a coupling between physical and operational times (γ -stable process and its inverse, respectively) is directed by an independent inverse tempered α -stable process. The coupling leads to two random processes underestimating and overestimating the real time t :

$$T_\gamma^-[S_\gamma(t)] < t < T_\gamma[S_\gamma(t)] \quad \text{for } t > 0 , \quad (4)$$

where $T_\gamma(\tau)$ is the γ -stable process, $S_\gamma(t) = \inf\{\tau : T_\gamma(\tau) > t\}$ is its inverse, and $T_\gamma^-(\tau) = \lim_{x \rightarrow \tau^-} T_\gamma(x)$ is the left-limit γ -stable process. The random

process $X_\gamma^-(t) = T_\gamma^-[S_\gamma(t)]$ has finite moments of any order whereas $X_\gamma(t) = T_\gamma[S_\gamma(t)]$ has even no the first moment (see details in [19]).

We take into account the process with the finite moments only, i.e., $X_\gamma^-(t) = T_\gamma^-[S_\gamma(t)]$ with $0 < \gamma \leq 1$. Its probability density is of the form

$$p^-(y, t) = \frac{\sin \pi \gamma}{\pi} y^{\gamma-1} (t - y)^{-\gamma}, \quad 0 < y < t. \quad (5)$$

Its moments of any order read

$$\begin{aligned} \langle X_\gamma^- \rangle &= \gamma t, \quad \langle (X_\gamma^-)^2 \rangle = \frac{\gamma(1+\gamma)}{2} t^2, \dots, \\ \langle (X_\gamma^-)^n \rangle &= \frac{\gamma(1+\gamma) \dots (\gamma+n-1)}{n!} t^n. \end{aligned}$$

Process $X_\gamma^-(t) = T_\gamma^-[S_\gamma(t)]$ is 1-similar and evolves to infinity as the real time t .

To obtain the widely observed fractional two-power relaxation pattern (1) we construct a compound subordinator which combines two following random processes: one is the coupling process $X_\gamma^-(t)$, and another is the inverse tempered α -stable process $Q_\alpha(t)$. The process $X_\gamma^-(t)$ directed by $Q_\alpha(t)$ is just the new compound subordinator, namely $W_{\alpha,\gamma}(t) = X_\gamma^-(Q_\alpha(t))$. We assume that $X_\gamma^-(t)$ and $Q_\alpha(t)$ are independent on each other. In this case the anomalous diffusion is determined by the following relation

$$p^r(x, t) = \int_0^\infty \int_0^\infty p^Y(x, y) p^-(y, \tau) g_\alpha(\tau, t) dy d\tau, \quad (6)$$

where $p^r(x, t)$ is the probability density of the subordinated process obtained from Y directed by $W_{\alpha,\gamma}(t)$, and $p^Y(x, y)$ is the probability density of the parent process Y . The explicit form of the latter is not important here. For simplicity, but without loss of generality, as the parent process can be taken the standard Brownian motion. The process $X_\gamma^-(t)$ is the essence of time clustering in the compound subordination. This process is approximated by a simple continuous-time random walk in which each waiting time is exactly equal to the jump. Consequently, a walker, moving along a Brownian trajectory in presence of the subordination by $X_\gamma^-(t)$ (without any government of $Q_\alpha(t)$), stops from time-to-time and overjumps through intermediate positions in the Brownian trajectory. In another words the process $X_\gamma^-(t)$ leads to a random partition (or clustering) of walker's trajectories.

3. Derivation of relaxation function

The primary, commonly accepted, interpretation of relaxation phenomena is based on the concept of exponentially relaxing objects (for example, dipoles) with different relaxation rates [13]. If the relaxing objects do not interact with each other, then their macroscopic relaxation function is described by the D law $\phi(t) = \exp(-\omega_p t)$. But the simplest situation, as it is known from many experiments, is realized very rarely. Since the relaxing objects can interact with their environment, their evolution to an equilibrium state has a complex (random walk-like) behavior which, in general, does not yield the D law. This interaction may be taken into account with an aid of the temporal subordination [6, 23]. In this case the relaxation function reads

$$\phi(t) = \int_0^\infty e^{-\omega_p \eta} p(\eta, t) d\eta,$$

where $p(\eta, t)$ is the subordinator pdf. Particularly, when such a subordinator is the inverse α -stable process $S_\alpha(t)$, the relaxation function takes simply the Mittag-Leffler (or the corresponding CC) form. Note that the solutions of fractional differential equations are just expressed in terms of the Mittag-Leffler function and its various generalizations (see, for example, the excellent books [24, 25]). They have extensive modern-day applications in the study of complex systems, which maintain the long-memory and nonlocal properties of the corresponding dynamics such as anomalous diffusion [26] and non-exponential relaxation [27, 28, 29, 30]. In particular, multi-particle systems with impact phenomena, that exhibit interactions between particles, have fractional dynamics [31]. Nevertheless, the fractional differential equations are only a macroscopic description of the complex system evolution, and their microscopic dynamics should be found out from other reasons.

If the exponentially decayed states are subordinated by the random process $W_{\alpha, \gamma}(t)$, then the time-domain relaxation function is expressed in terms of the integral relation

$$\begin{aligned} \phi(t) &= \int_0^\infty \int_0^\infty e^{-\omega_p y} p^-(y, \tau) g_\alpha(\tau, t) dy d\tau \\ &= \frac{\sin \pi \gamma}{\pi} \int_0^\infty \int_0^1 e^{-\omega_p \tau z} z^{\gamma-1} (1-z)^{-\gamma} g_\alpha(\tau, t) dz d\tau, \end{aligned} \quad (7)$$

where ω_p is a constant. For the experimental study the frequency-domain

representation of the latter function

$$\varphi^*(\omega) = \int_0^\infty e^{-i\omega t} \left(-\frac{d\phi(t)}{dt} \right) dt \quad (8)$$

is of more interest. It is well known [13, 14] that the complex dielectric susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega) \propto \varphi^*(\omega)$ of most relaxing substances shows a peak in the loss component $\chi''(\omega)$ at a characteristic frequency ω_p . After integration of Eq.(8) by means of Eq.(7) we get

$$\varphi^*(\omega) = 1 - \left(\frac{(i\omega/\omega_p + \sigma)^\alpha - \sigma^\alpha}{1 - \sigma^\alpha + (i\omega/\omega_p + \sigma)^\alpha} \right)^\gamma, \quad (9)$$

where $0 \leq \sigma = \delta/\omega_p < \infty$ is a positive constant. Such a frequency-domain relaxation function fits the asymptotic behavior of the dielectric susceptibility $\chi(\omega)$ with different low- and high-frequency power tails, namely

$$\begin{aligned} \Delta\chi(\omega) &\propto (i\omega/\omega_p)^\gamma & \text{for } \omega \ll \omega_p, \sigma \neq 0, \\ \chi(\omega) &\propto (i\omega/\omega_p)^{-\alpha} & \text{for } \omega \gg \omega_p. \end{aligned} \quad (10)$$

The main feature of the susceptibility obtained above is that the indices $m = \gamma$ and $1 - n = \alpha$ are determined by different processes. The index α is connected with the tempered α -stable process only, and γ is basically determined by the coupling process $T_\gamma^-[S_\gamma(t)]$. Thus, by the law (9) we can describe any fractional two-power relaxation dependency with both $m > 1 - n$ and $m < 1 - n$ (see Fig. 2). This advantage allows one to get any point on the relaxation diagram shown in Fig. 1, but here is some nuances.

Recall that if one uses the conventional α -stable process instead of the tempered one, then in order to come to both fractional two-power relaxation dependencies, both undershooting $X_\gamma^-(S_\alpha(t))$ and overshooting $X_\gamma(S_\alpha(t))$ processes in the corresponding compound subordinator should be considered [1]. In particular, the overshooting process clearly leads to the HN law of relaxation, but the process itself has no finite moments. Notice that the ordinary α -stable process influences on both indices (m and $1 - n$) in the response function (see Table 2), whereas the undershooting process acts only on the index m , and the overshooting process changes only the index $1 - n$. As the indices $0 < \alpha, \gamma < 1$, the value $\alpha\gamma < \alpha$ holds true always. With the parameter $\delta = 0$ the tempered α -stable process becomes simply α -stable, and the frequency-domain relaxation response $\varphi^*(\omega)$ coincides with the form

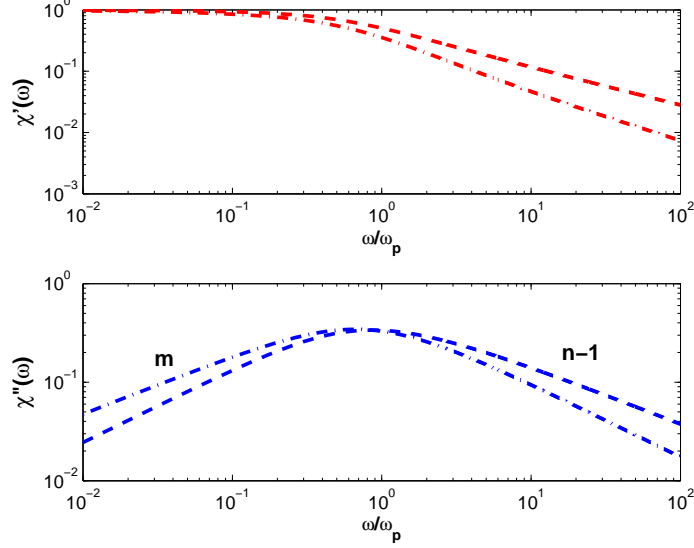


Figure 2: (Color online) Real and imaginary terms of susceptibility $\chi(\omega)$ (9), showing the power-law property (1). Dashed lines represent $m = \gamma = 0.75$ and $1 - n = \alpha = 0.6$ whereas dash-dotted lines correspond to $m = \gamma = 0.6$ and $1 - n = \alpha = 0.75$. The parameter σ equals to 0.5.

arising from the aforesaid undershooting process in the corresponding compound subordinator [32]. In this case the low-frequency asymptotics $\Delta\chi(\omega)$ is proportional to $(i\omega/\omega_p)^{\gamma\alpha}$. The CC law is just obtained from Eq.(7) for $\sigma = 0$ and $\gamma = 1$. The CC plot for Eq. (9) is shown in Fig. 3.

When the parameter γ is equal to 1, the expression (9) simplifies to the form characteristic for the ordinary tempered relaxation. This is not surprising because in this case the probability density $p^-(y, \tau)$ becomes the Dirac-delta function $\delta(\tau - y)$. When the parameter α tends to 1, the probability density $g_\alpha(t, \tau)$ reads as $\delta(t - \tau)$. For $\delta = \alpha = 1$ we obtain clearly the ordinary exponential relaxation. Moreover, the real and imaginary parts of the permittivity data as a function of frequency obtained for the investigated samples of $\text{Cd}_{0.99}\text{Mn}_{0.01}\text{Te:Ga}$ at various temperatures are well fitted by the dependence (9) with the parameters $\sigma \approx 0$, $\alpha \approx 1$ and $\gamma \approx 0.63$ [33]. This dependence is also applicable for appropriate glass-forming materials such as, for example, ones studied in [14, 34].

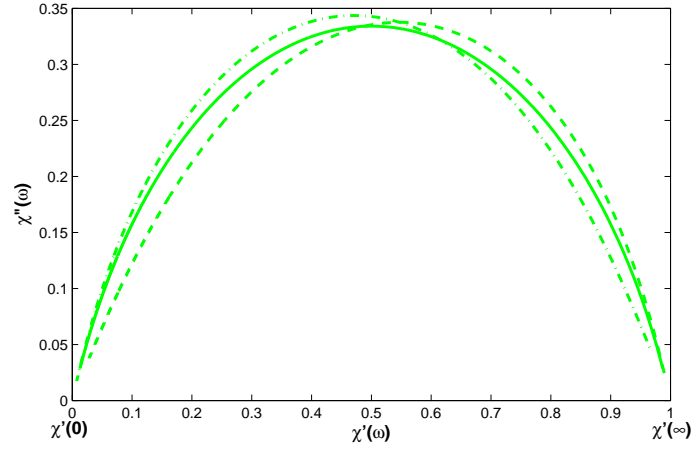


Figure 3: (Color online) Cole-Cole plot for Eq. (9) with the same values of parameters as in Fig. 2: the solid (symmetric) line corresponds to the CC susceptibility with $m = 1 - n = 0.75$ and $\delta = 0$, the dashed (asymmetric) line relates to the tempered case for $m > 1 - n$ whereas the dash-dotted line (asymmetric in another side) represents the tempered case for $m < 1 - n$.

Table 2: Asymptotic behavior of the frequency-domain response function for fractional relaxation scenarios (see also [1]).

Type of relaxation	Operational time (subordinator)	m	$1 - n$
Mittag-Leffler (ML)	$S_\alpha(t)$	α	α
Havriliak-Negami	$X_\gamma(S_\alpha(t))$	α	$\alpha\gamma$
Generalized Mittag-Leffler (GML)	$X_\gamma^-(S_\alpha(t))$	$\alpha\gamma$	α
Tempered response (9)	$X_\gamma^-(Q_\alpha(t))$	γ	α

4. Conclusions

This approach gives a chance to include the fractional two-power-law dependency, widely known from relaxation experiments, with $m > 1 - n$ and $m < 1 - n$ in the framework of tempered relaxation. Before, with this in mind to describe the diagram of Fig. 1, the HN relaxation law was completed for $m < 1 - n$ by the GML (Generalized Mittag-Leffler) dependence (see more details in [1, 35]). Although the new form of the frequency-domain relaxation function is more complicated than the HN's law, the latter requires necessarily to account for the GML relaxation law because a considerable part of experimental data in relaxation studies cannot be described by any modified version of the HN law. The proposed attempt also permits one to select easily effects influencing on the power character of low- and high-frequency tails in the relaxation function. The appearance of the passage from $m < 1 - n$ to $m > 1 - n$ and vice versa in the same material reflects structural changes to show what has a dominant character, either flip-flops of dipoles (relaxing objects) at short times or cluster patterns of dipole sets at long times. The short-time behavior can be produced by traps, and the long-time trend is a result of long-range interactions of dipoles via their cluster and collective regions. It should be stressed that in this scheme we use only the random processes with finite moments what is important for analysis and interpretation of experimental data [14].

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